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Evaluation of recharge areas of Arusha Aquifer, Northern Tanzania: application of water isotope tracers

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ABSTRACT

In Arusha urban, northern Tanzania, groundwater contributes about 80% of the water supply. However, elevated fluoride levels and evidence of anthropogenic pollution have been reported in the groundwater around Mount Meru which is a water source for Arusha urban. This study aims at understanding the recharge areas and flow pathways of groundwater in what has been a poorly monitored area. The study uses the isotopic ratio of oxygen and hydrogen to estimate the groundwater recharge area and flow pathway. The results show the recharge elevation of groundwater is between 1,800 and 3,500 m above mean sea level on the slopes of Mount Meru. The average fluoride contents in the study area are 5.3 ± 0.4 mg/L greater than the limits of 1.5 mg/L set by the World Health Organization (WHO) and Tanzania. The nitrate concentration of 83.9 mg/L at the lower elevation areas (<1,400 m above mean sea level) exceeds the 50 mg/L WHO limit. The relationship of F^- with $\delta^{18}O$ and NO_3^- suggests the leaching of fluoride in high altitudes and dilution in lower altitudes.

Key words | altitude effect, Arusha Aquifer, groundwater, isotopes, recharge area, Tanzania

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HIGHLIGHTS

- Management of groundwater resources is essential to maintain the supply of freshwater.
- Adequate and precise demarcation of the groundwater area for protection is critical.
- Isotopic ratio of oxygen and hydrogen is used to estimate the recharge area and flow pathway.
- Meteoric water is the source of groundwater recharge in Mount Meru watershed.

INTRODUCTION

Shortage of adequate surface water sources, urban development, escalating per capita water consumption, and the influence of climate change have made groundwater the main source of water for domestic uses in many urban areas around the globe, including Tanzania (Li 2013; Foster Bousquet & Furey 2018). However, groundwater sources are often faced with various dynamics such as geochemistry, topography, geology, water–rock interaction, and anthropogenic activities (Mduma *et al.* 2016). In Arusha city, northern Tanzania, groundwater from wells and springs provides more than 80% of the freshwater used for both domestic and industrial purposes (Chacha *et al.* 2018a). However, the greatest threats to maintaining freshwater supply in the city are the prevalence of fluoride contamination, a decrease in groundwater reserve, and degradation of water quality due to human activities in the potential recharge areas

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(Chacha *et al.* 2018a). A comprehensive examination of the recharge areas and flow pathway of groundwater is a significant tool when considering sustainable groundwater resources management (Nakaya *et al.* 2015).

The Arusha urban water authority has recently developed an additional well field with nine boreholes for domestic uses (≥200 m deep) at Ngaramtoni area on the south-western slopes of Mount Meru, on the north-western part of Arusha city. The new well-field construction demonstrates a commitment to long-term reliance on the Arusha Aquifer. However, groundwater resources in the area are not adequately protected and anthropogenic activities have been reported in the recharge areas recently (Chacha *et al.* 2018a). A precise demarcation of the areas of protection is critical (Nayak 2016). To achieve this, identification of the spatial distribution of the predominant recharge areas and flow pathway of groundwater is needed (Carrillo-Rivera & Varsányi 2007; Nayak 2016). The present study aimed to identify the spatial distribution of the predominant recharge areas of groundwater used in Arusha urban and to understand the general flow pathway of groundwater for fluoride contamination in Arusha urban groundwater resources.

Several methods such as multivariate statistical analysis using chemical tracers, etc. (Bakari *et al.* 2012) can be used to delineate the recharge areas of groundwater. However, the stable isotopic ratio of oxygen and hydrogen ($\delta^{18}O$ and δD) is the best method for tracing flow pathways and clarifying the groundwater origin and source areas (Nakaya *et al.* 2015) because these isotopes are naturally contained in the water molecule and cannot be modified by water–rock contacts (Kim 2007). Furthermore, the isotopic altitude effect is very useful for tracing and distinguishing groundwater recharged at high altitudes from that recharged at low altitudes (Li 2013; Nakaya *et al.* 2015). Various researchers have successfully used the isotopic altitude effect of the isotopic ratio of oxygen to determine the recharge areas and flow pathway of groundwater (e.g., Li 2013; Nakaya *et al.* 2015). Bouchaou *et al.* (2009) reported that the variation in the isotopic values of groundwater is influenced by differences in the altitude of recharge areas. As the mean annual air temperature becomes low at high elevations, the composition in the stable isotope of water vapor in the atmosphere decreases by isotope fractionation (Farid *et al.* 2014).

Therefore, this study applied the stable isotope ratio method on groundwater and surface water samples from Mount Kilimanjaro watershed at an altitude >1,500 m.a.s.l. The results of the study primarily provide critical information for the management of the groundwater resources in the study area. They might also help to indicate what could be happening in other areas and how it can best be studied.

THE STUDY AREA

The present study was conducted in urban areas of Arusha (including the Arusha district council) located on the south-western slopes of Mount Meru in the north-eastern part of Tanzania with an elevation of 1,400 m.a.s.l. (Figure 1). According to the Tanzania population and housing census data of 2012, the approximated total population of the area is 739,640 (NBS 2013).

The area experiences a tropical climate with dry and wet seasons and the rainfall pattern is bimodal, with short rains between November and December and long rains between March and May or June with a total average annual precipitation of 842 mm (Chacha *et al.* 2018b). The maximum temperature varies between 13 and 30 °C with an annual mean value of 25 °C and the area is characterized by a narrow variation of relative humidity (55–75%) with 924 mm annual potential evapotranspiration (Chacha *et al.* 2018b).

The area is studded by volcanic deposits of variable ages and dumped alluvial residues (Ghiglieri *et al.* 2010). According to Chacha *et al.* (2018b), Mount Meru is the focal point of volcanic events in the area and the lava flow forms the main volcanic rocks such as basalts to phonolitic and nephelinitic tuff. These act as an aquitard, which restricts the infiltration of groundwater, and directs the surface run-off toward the lower slopes. Faults and fractures formed due to volcanic and tectonic activities act as groundwater conduits. Moreover, the study area is described by volcanic and sedimentary hydrogeological formation composed of rocks with minerals such as fluorapatite, natrite, halite, calcite, chabazite, nepheline, biotite, and illite. The geological properties in the area change with geologic time and the main groundwater aquifer is characterized by volcanic ash, pyroclastic deposits, weathered and fractured materials

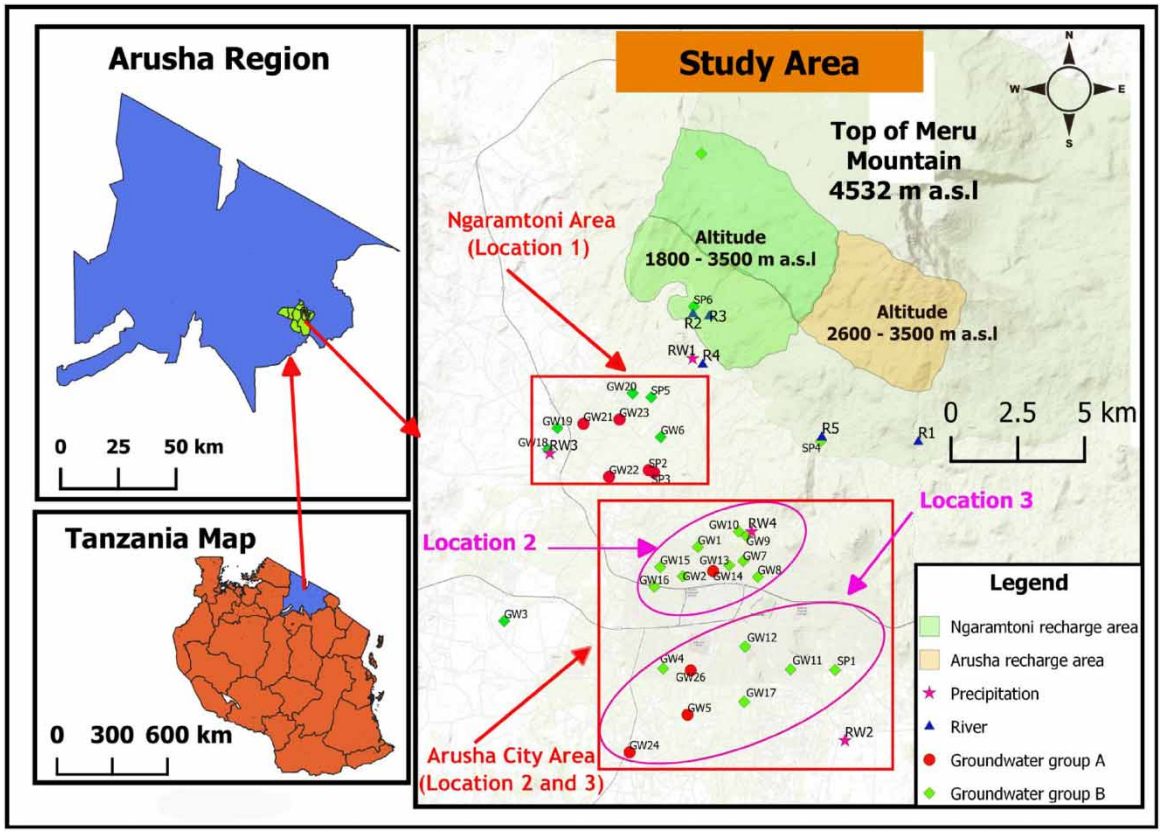

Figure 1 | Map of the Arusha region and details of the study area.

such as basalts, and phonolitic to nephelinitic materials (Ghiglieri *et al.* 2010; Chacha *et al.* 2018b).

MATERIAL AND METHODS

Sampling and analytical measurements

Groundwater and river water samples were collected throughout the study area (Figure 1) in September 2018 for groundwater and from March to May 2019 for river water to document the spatial disparity in the isotopic signature of $\delta^{18}O$ and δD. The groundwater samples ($n = 32$), as presented in Table 2, were collected from springs as well as from public and privately owned deep and shallow wells close to the points of discharge to reduce the influence of evaporation and pollution from the atmosphere. The sampling points were as mapped in Figure 1 at altitudes ranging from 1,299 to 1,904 m.a.s.l. in Mount Meru watershed.

The depth of the sampling wells from the ground surface ranged from 48 to 273 m for public wells and 11 to 26 m for privately owned wells. Most of the public wells were labeled with the wells' information, but the privately owned wells were not labeled and information was obtained from the owners. The groundwater levels were measured using a water level meter. The depth to groundwater level ranged from 2.7 to 93.2 m for public wells and 1.98 to 7.03 m for privately owned wells. One artesian well was also observed during the field survey.

The primary sources of groundwater recharge in the study area are precipitation and rivers. River water samples were collected from different points based on the ease of access within the study area to establish the isotopic composition of different recharge sources. To develop the local meteoric water line (LMWL) for Mount Meru watershed, rainwater samples were collected from five locations distributed in altitudes between 1,294 and 1,813 m.a.s.l., as presented in Figure 1. Also, the precipitation information

of Dar es Salaam and Dodoma as presented in Table 2 were obtained from the International Atomic Energy Agency (IAEA) database. To trace the flow pathways and recharge areas of groundwater, water samples from ($n = 26$) wells, ($n = 6$) springs, ($n = 5$) rivers, as well as rainwater samples ($n = 4$) from various altitudes, were collected for measuring stable isotopic ratios of $\delta^{18}O$ and δD.

The recharge area of groundwater can be estimated by using δD and $\delta^{18}O$ values and show their relationship to the altitude at which precipitation might have infiltrated the groundwater. The altitude effect with a gradient of −0.13‰ per 100 m was applied to calculate the recharge elevation of groundwater because the composition of stable isotope decreases with a rise in altitude since the mean annual air temperature becomes low at high elevation (Farid *et al.* 2014).

Measurements of temperature, electrical conductivity (EC), pH, oxidation–reduction potential (ORP), and dissolved oxygen (DO) for well, spring, and river water samples were performed onsite. The sampling locations and respective elevations were recorded using a hand-held global positioning system (GPS) receiver. Precipitation samples were collected through a high-density polyethylene (HDPE) funnel (30 cm in diameter) into 5 L plastic containers which were covered with aluminum foil and buried in the soil at open sky locations to minimize evaporation (Bakari *et al.* 2012).

All water samples for stable isotopic ratios and chemical composition were collected in 50 mL pre-cleaned HDPE bottles filtered through a 0.2 μm membrane filter. To ensure that the samples represented the groundwater at the location, each well and spring was purged before sampling until the monitoring values of pH, EC, ORP, DO and temperature stabilized. The sampling bottles were rinsed three times with the water samples and sealed tightly to avoid contamination and loss by evaporation.

The laboratory analyses were undertaken at Shinshu University in Japan. The isotopic composition values for δD and $\delta^{18}O$ were measured using $\delta D/\delta^{18}O$ Isotopic Water Analyzer (Picarro L2130-i) (Nakaya *et al.* 2015) with respective analytical precision of 0.1‰ and 0.02‰. The isotopic results were recorded with respect to Vienna Standard Mean Ocean Water (V-SMOW) in δ notation. The major anions and cations were analyzed using non-suppressed ion chromatography with IC-C4 and IC-A3 columns (Shimadzu) and a detection limit of <0.1 mg/L. The measurement of alkalinity was undertaken by titration against 0.02 N HCl using a mixture of bromocresol green and methyl red (BCG-MR) as an indicator (pH 4.8).

RESULTS AND DISCUSSION

Hydrogeochemistry

Table 2 shows the physicochemical parameters and isotopic composition of $\delta^{18}O$ and δD for groundwater, river water, and precipitation samples. The hydrogeochemistry analysis using the Piper diagram shows that the Na–HCO_3 water type dominates for both well, spring, and river waters except sample SP1 from spring which is the Na–Ca–HCO_3 water type (Figure 2). The pH of the groundwater samples ranged between 6.18 and 8.56 with a mean value of 7.1 ± 0.09, indicating that the water was weakly acidic to alkaline, while pH values for the river water varied between 6.8 and 8.2. About 44% of all the analyzed groundwater samples were weakly acidic (pH < 7). A similar observation from weakly acidic to alkaline groundwater in Mount Meru watershed has been reported (Elisante & Muzuka 2016; Mduma *et al.* 2016; Chacha *et al.* 2018b).

The temperature of the groundwater samples varied between 17.2 and 25.5 °C with a mean value of 21.1 ± 0.4 °C, while that of river water samples varied between 13.4 and 18.2 °C with a mean value of 15.9 ± 0.9 °C. The EC of the groundwater samples varied widely, from 16.19 to 172.80 mS/m with a mean value of 70.6 ± 6.5 mS/m.

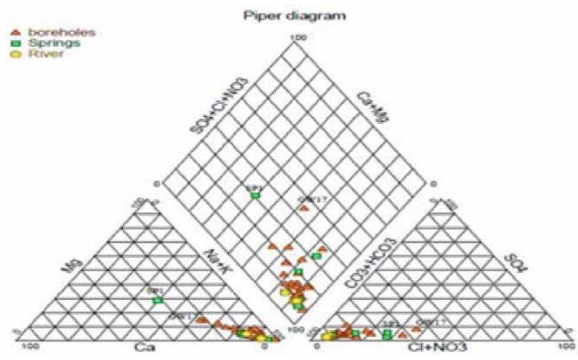

Figure 2 | Classification of water chemistry in the Piper plot.

However, the EC of river waters ranged within that of groundwater (16.19–54.50 mS/m), indicating stronger water–rock interaction in groundwater relative to river water. The concentrations of Cl^- and Na^+ suggest the dissolution of ions from rocks, which is also reflected in the EC (Table 1a), as also reported in other parts of the globe (Li 2013).

Generally, the study area had elevated fluoride concentrations in both groundwater and river water. The fluoride concentration in the groundwater varied widely from 1.23 to 10.09 mg/L, with an average value of 5.13 ± 0.4 mg/L. Similar ranges of fluoride concentrations in naturally contaminated geothermal waters located in volcanic areas have been reported in other parts of the world (Edmunds & Smedley 2013). Likewise, the stable isotopic ratios of oxygen and hydrogen show that the groundwater in the present study area is of meteoric origin (rain and/or snow), though elevated fluoride contamination suggests mixing with geothermal waters. River water samples were observed to have a slightly narrower range of fluoride concentration (2.1–9.3 mg/L) with an average value of 5.9 ± 1.3 mg/L. The level of fluorides in about 69% of all the analyzed groundwater samples exceeded the recommended upper limit for drinking water of 1.5 mg/L set by both WHO and Tanzania.

The fluoride concentrations further showed correlations with Na^+ ($r = 0.70$, $P < 0.05$) and alkalinity ($r = 0.60$, $P < 0.05$) as shown in Table 1a, suggesting that like Na^+, the fluoride contamination is natural through water–rock contact mainly from volcanic rocks. These findings are also in line with the suggestions by Kim & Kim (2011) for the relationships of fluorides with Na^+ and alkalinity in groundwater.

The results presented in Table 2 show a wide range of NO_3^- contents in the groundwater samples. The lowest NO_3^- level of 0.7 mg/L and the highest NO_3^- level of 83.9 mg/L were observed at altitudes of 1,447 and 1,341 m.a.s.l., respectively. Compared to the WHO guidelines for drinking water of 50 mg NO_3^- per liter, about 9.4% of all analyzed groundwater samples showed higher NO_3^- concentrations. The SO_4^{2-} contents in the groundwater samples ranged from 2.17 mg/L at the altitude of 1,760 m.a.s.l. to 106 mg/L at the altitude of 1,343 m.a.s.l. with an average value of 18.7 ± 3.5 mg/L, whereas in river water samples, the SO_4^{2-} concentrations ranged from 1.6 to 9.5 mg/L. The presence of SO_4^{2-} could be due to the dissolution of sulfide deposits in the soil layer or anthropogenic pollution from farming activities in the lower altitude parts of the study area. Recent studies (Elisante & Muzuka 2016; Mduma *et al.* 2016) have similarly reported elevated levels

Table 1 | Statistical correlation between chemical compositions of groundwater (a) and the relationship of well depths, recharge altitudes, sampling altitudes and isotopic composition of groundwater (b)

	Cl^- (mg/L)	Na^+ (mg/L)	EC (mS/m)	F^- (mg/L)	NO_3^- (mg/L)	$\delta^{18}O$ (‰)	Alkalinity meq/L
(a)							
Cl^- (mg/L)	1						
Na^+ (mg/L)	0.63	1					
EC (mS/m)	0.8	0.96	1				
F^- (mg/L)	0.08	0.70	0.52	1			
NO_3^- (mg/L)	0.53	0.07	0.21	−0.2	1		
$\delta^{18}O$ (‰)	0.62	0.12	0.3	−0.38	0.2	1	
Alkalinity (meq/L)	0.61	0.9	0.96	0.60	−0.01	0.2	1
	Well depth (m)	**Recharge altitude (m)**	**Sampling altitude (m)**	**$\delta^{18}O$ (‰)**			
(b)							
Well depth (m)	1						
Recharge altitude (m)	0.73	1					
Sampling altitude (m)	0.72	0.66	1				
$\delta^{18}O$ (‰)	−0.73	−1	−0.66	1			

of NO_3^- and SO_4^{2-} in groundwater sources in the lower slopes of Mount Meru.

The lowest Cl^- concentration 2.0 g/L (fluoride = 2.1 mg/L) in groundwater samples was noted at a high altitude of 1,760 m.a.s.l., whereas the highest Cl^- concentration of 64.4 mg/L (fluoride = 2.2 mg/L) was observed at a lower elevation of 1,320 m.a.s.l (Table 2), suggesting the recharge and discharge areas of the aquifer, respectively. These observations are in line with earlier reports (e.g., Dattaa *et al.* 1996). There is no correlation between fluoride and Cl^- in our water samples (Table 1), indicating that Cl^- is likely to come from anthropogenic influences (Bouchaou *et al.* 2009; Olaka *et al.* 2016; Makoba & Muzuka 2019).

The high NO_3^- concentrations in the groundwaters could be an indicator of anthropogenic pollution (Kim & Kim 2011). Similar studies by Krishnaraj *et al.* (2011) also report that higher Cl^- and NO_3^- contents in groundwaters indicate anthropogenic contamination from domestic wastewater, animal manure, and application of fertilizers. Slightly elevated SO_4^{2-}, NO_3^-, and Cl^- contents of up to about 106, 83.9, and 64.4 mg/L, respectively, in the groundwater could most likely be due to anthropogenic activities since they were observed in shallow wells (≤100 m deep), located in the informal settlements of Arusha city which are described by a high population density and poor sanitation facilities. The results thus suggest local recharges to shallow groundwater, especially in the topographically low parts of the study area.

The water sample WG25 collected from the shallow well (11 m deep) was observed to have negligible NO^{3-} (2.3 mg/L) and elevated Cl^- (50.7 mg/L) contents, indicating groundwater discharging conditions. The upward moving groundwater from the subsurface is fundamentally free of NO^{3-} and has high contents of Cl^- due to excess evaporation. However, high Cl^- can also represent the mixing of polluted water from human activities (Carrillo-Rivera & Varsányi 2007). Moreover, according to Koh *et al.* (2012), the relationship of NO^{3-} and Cl^- contents as shown in Table 1a reveals that a significant portion of Cl^- comes from anthropogenic sources.

The Na^+ concentration showed a wide range from 28 to 341.1 mg/L with a median value of 121 mg/L. The Ca^{2+} contents range from 1.4 to 43.1 mg/L, while the concentration of K^+ and Mg^{2+} varied from 8.7 to 69.0 and 0.6 to 16.8 mg/L, respectively. The present study observed high Ca^{2+} contents at a lower altitude of Mount Meru watershed.

It should be noted that the statistically significant positive relationship between well depth and elevation of sampling location ($r = 0.72$, $P < 0.05$), as presented in Table 1b, indicates that most wells at higher altitudes are deep.

Stable isotopes of water

The stable isotopic values of the groundwater samples ranged from −6.0‰ to −4.0‰ for $\delta^{18}O$ and from −31.3‰ to −17.7‰ for δD, whereas the isotopic composition of the river water samples varied from −6.3‰ to −4.9‰ for $\delta^{18}O$ and from −34.7‰ to −23.0‰ for δD (Table 2). These results show that the river water samples had relatively narrow isotopic ranges and were more isotopically depleted than the groundwater samples, an indication of elevation effects since river water was sampled from high elevation areas (>1,700 m a.s.l) on slopes of Mount Meru (Table 2).

Similarly, the ratio between the isotopic values of $\delta^{18}O$ and δD varied locally due to climatic and geographical differences in the area. Thus, they represent the LMWL. This study estimated the LMWL for Tanzania ($\delta D = 7.037\delta^{18}O + 7.051$) using the precipitation data from the IAEA website recorded at Dar es Salaam and Dodoma stations. Dar es Salaam station is located in the coastline (latitude 6.88_S, longitude 39.20_E, 55 m.a.s.l.), and according to Rozanski *et al.* (2013), this is the only place within the global network of isotopes in precipitation (GNIP) in East Africa that records $\delta^{18}O$ values of rainfall directly from the Indian Ocean. Other studies (e.g., Mckenzie *et al.* 2010) used a virtually similar LMWL ($\delta D = 7.057\delta^{18}O + 7.0$) in related studies on Mount Kilimanjaro, which was developed by using the precipitation data obtained from the IAEA, recorded from 1960 to 1976.

Figure 3 shows the $\delta^{18}O$ and δD values of the groundwater, precipitation and river waters in reference to the global meteoric water line (GMWL) expressed as $\delta D = 8\delta^{18}O + 10$ (Levin *et al.* 2009; Rozanski *et al.* 2013), LMWL for Tanzania, and an average of meteoric water lines for Africa defined by Levin *et al.* (2009) as $\delta D = 7.48\delta^{18}O + 10.1$. The linear equation resulting from the $\delta D - \delta^{18}O$ relationship is expressed as $\delta D = 7.3\delta^{18}O + 11.41$. The LMWL for the study area revealed a slope (=7.3) which is virtually identical

Table 2 | Physico-chemical and isotopic composition of groundwater, river water and precipitation samples

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Oltulelei	Borehole	GW1	1481	180	6.8	19.7	44.2	3.85	87.5	18.8	3.9	6.9	4.1	8.5	18.4	8.0	−5.2	−26.8
Ilkiurei	Borehole	GW2	1430	100	7.31	21.4	50.1	5.05	123.1	17.9	1.9	6.5	5.3	13.3	37.8	9.3	−5.1	−24.9
Magereza	Borehole	GW3	1369	130	7.21	24.4	111	10.80	220.4	45.3	9.2	9.3	3.8	31.9	25.6	26.0	−4.9	−25.3
Sombetini shuleni	Borehole	GW4	1341	100	7.04	22.5	69.3	4.36	125.5	30.0	4.9	15.2	3.6	34.5	83.5	12.9	−5.0	−24.8
Longdong	Borehole	GW5	1314	–	7.19	23.9	98.7	7.72	180.4	39.0	9.3	28.5	5.2	53.2	42.3	24.2	−4.4	−21.7
Seedfarm No.6	Borehole	GW6	1623	273	7.67	17.2	43.2	3.70	90.8	17.4	1.1	7.6	6.1	5.0	6.5	7.2	−5.6	−27.7
Old Sanawari	Borehole	GW7	1460	88.4	7.26	21.3	41.2	2.63	55.6	16.1	6.0	13.7	2.5	11.4	19.6	11.3	−4.7	−22.4
Moivo II	Borehole	GW8	1449	103.5	7.49	21.1	36.5	3.33	71.4	12.6	3.6	5.8	2.2	5.0	4.4	3.3	−4.6	−21.1
Loruvani yard	Borehole	GW9	1501	81	7.24	20.9	35.8	3.27	61.5	17.2	4.3	10.0	3.5	5.6	11.9	7.6	−4.7	−22.1
Loruvani bondeni	Borehole	GW10	1499	63	7.37	20.3	34.7	3.20	72.9	15.5	1.7	4.1	5.0	4.8	5.8	5.5	−5.2	−25.6
Moivo I	Borehole	GW11	1349	–	7.25	22.1	39.6	3.10	51.1	16.4	7.2	17.6	2.1	10.8	20.1	4.9	−4.4	−20.3
Machale	Spring	SP1	1322	–	6.68	21.7	34.3	2.26	28.1	8.7	12.7	23.0	1.2	12.5	34.0	5.2	−4.5	−20.3
EMCO	Borehole	GW12	1369	78	7.54	21.7	42.9	4.34	94.0	20.5	5.7	5.2	3.8	10.7	1.9	6.2	−4.7	−21.6
Ilkiloriti	Borehole	GW13	1451	181.5	7.23	21.7	39.4	3.20	81.9	15.9	2.2	4.2	5.5	6.4	8.0	7.8	−4.9	−23.2
Mianzini	Borehole	GW14	1447	141.5	8.56	22.3	52.4	4.62	127.9	10.1	0.6	1.4	8.8	9.0	0.7	6.0	−5.0	−23.3
Kiranyi I	Borehole	GW15	1435	189	6.95	21.1	49.7	4.44	99.1	18.2	3.6	11.4	4.6	6.8	13.7	9.7	−5.2	−25.2
Sakina	Borehole	GW16	1418	91.4	7.24	21.7	47.5	4.15	103.2	15.7	2.1	6.7	6.1	7.0	9.8	8.2	−5.3	−26.8
Lemala	Borehole	GW17	1320	48	6.97	23.7	88.5	4.81	118.8	31.0	16.8	43.1	2.2	64.4	83.9	33.9	−4.2	−19.6
Monduli	Borehole	GW18	1500	120	6.82	22.3	90.9	8.32	175.5	34.1	5.4	22.6	5.7	16.7	15.1	27.8	−5.2	−27.9
Shamba la mbegu	Borehole	GW19	1514	–	6.87	22.0	74.8	7.40	136.6	35.3	5.7	19.7	4.5	14.0	6.1	18.7	−5.1	−26.0
Saida School	Borehole	GW20	1641	–	6.66	18.0	87	8.00	187.2	38.9	1.7	11.6	6.3	8.3	21.7	19.1	−5.6	−30.1
Mzee Ally	Borehole	GW21	1542	–	6.76	17.6	136.1	12.82	282.5	58.6	5.9	29.5	8.8	26.0	29.0	41.9	−5.1	−26.9
Levolosi juu	Spring	SP2	1594	–	6.36	20.9	76.5	6.24	128.8	49.8	5.3	19.2	5.3	18.6	38.4	21.0	−4.6	−23.8
Levolosi chini	Spring	SP3	1582	–	6.18	20.8	97.2	5.78	144.9	52.0	4.8	20.3	5.5	36.2	77.0	25.4	−4.6	−24.1
Seliani	Borehole	GW22	1536	–	6.32	21.4	93.2	8.82	180.2	69.0	4.3	9.7	7.7	11.4	12.8	14.9	−4.8	−25.3
Mzee Zakaria	Borehole	GW23	1597	–	6.74	19.2	95.3	8.58	211.3	35.5	1.3	11.5	10.1	12.8	21.4	23.5	−5.5	−30.2
Muriet ofisi-kata	Borehole	GW24	1299	15	6.94	25.5	131.7	11.72	295.5	34.9	5.9	13.8	7.6	52.8	16.8	39.9	−4.8	−25.2
Unga Ltd 1	Borehole	GW25	1343	11	6.64	21.7	121.9	10.27	198.3	57.5	15.3	41.8	5.9	50.7	2.3	41.1	−4.0	−17.7
Unga Ltd 2	Borehole	GW26	1343	26	7.40	22.8	172.8	16.34	341.1	59.4	15.0	39.9	6.9	56.1	8.7	105.7	−4.0	−17.8
Themi	Spring	SP4	1760	–	7.65	18.2	16.19	1.44	28.5	10.2	1.1	3.4	2.1	2.0	1.2	2.2	−4.9	−23.0
Njoro	Spring	SP5	1662	–	7.6	19.8	52.2	4.65	108.5	25.2	0.9	6.4	7.3	6.0	10.1	11.0	−6.0	−31.3

(*continued*)

Table 2 | continued

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Oloshaa	Spring	SP6	1904	–	7.7	17.5	55.9	5.00	109.7	40.9	1.2	4.4	4.6	5.4	4.6	8.9	−5.3	−28.2
Nduruma	River	R1	1891	–	8.24	13.4	17.21	1.28	30.6	8.3	0.5	1.8	3.7	2.2	0.8	1.6	−5.70	−28.1
Emboocho chini	River	R2	1907	–	7.3	16.1	43.8	3.85	88.1	23.0	1.0	5.9	7.2	4.9	6.2	7.4	−6.15	−34.7
Emboocho juu	River	R3	1971	–	6.84	14.7	54.5	3.76	88.0	23.0	0.9	5.3	7.3	4.9	5.8	7.8	−6.16	−34.4
Seliani	River	R4	1803	–	7.55	17.2	49.6	4.10	95.7	25.9	1.1	6.0	9.3	6.2	2.9	9.5	−6.27	−34.0
Themi	River	R5	1760	–	7.65	18.2	16.19	1.44	28.5	10.2	1.1	3.4	2.1	2.0	1.2	2.2	−4.86	−23.0
Timbolo School	Rainfall	RW1	1813	–	–	–	–	–	–	–	–	–	–	–	–	–	−5.61	−27.9
Moshono-Laizer	Rainfall	RW2	1294	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.29	−6.4
NGAWASA BH	Rainfall	RW3	1490	–	–	–	–	–	–	–	–	–	–	–	–	–	−7.04	−42.4
Loruvani yard	Rainfall	RW4	1501	–	–	–	–	–	–	–	–	–	–	–	–	–	−6.34	−34.5
Dar es Salaam	Rainfall	196103	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.3	−0.6
Dar es Salaam	Rainfall	196104	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1	−13.7
Dar es Salaam	Rainfall	196105	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2	6.2
Dar es Salaam	Rainfall	196106	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.1	2.5
Dar es Salaam	Rainfall	196107	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.1	−29.2
Dar es Salaam	Rainfall	196108	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.1	1.8
Dar es Salaam	Rainfall	196109	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.6	5.6
Dar es Salaam	Rainfall	196110	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−5.1	−28
Dar es Salaam	Rainfall	196112	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−6.6	−45.2
Dar es Salaam	Rainfall	196201	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.3	−3.1
Dar es Salaam	Rainfall	196202	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.5	−10.6
Dar es Salaam	Rainfall	196203	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.7	6.8
Dar es Salaam	Rainfall	196204	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.6	−5.6
Daar es Salaam	Rainfall	196205	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.4	−3.8
Dar es Salaam	Rainfall	196206	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.2	4.4
Dar es Salaam	Rainfall	196207	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.4	1.2
Dar es Salaam	Rainfall	196208	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.2	−4.4
Dar es Salaam	Rainfall	196210	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.8	−4.4
Dar es Salaam	Rainfall	196211	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.7	−1.9
Dar es Salaam	Rainfall	196212	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.6	−6.9
Dar es Salaam	Rainfall	196301	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.4	−3.1

(*continued*)

Table 2 | continued

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dar es Salaam	Rainfall	196302	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2	−5.3
Dar es Salaam	Rainfall	196303	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.7	−16.8
Dar es Salaam	Rainfall	196304	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.4	−19.9
Dar es Salaam	Rainfall	196305	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.1	−0.6
Dar es Salaam	Rainfall	196306	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.4	−25.5
Dar es Salaam	Rainfall	196307	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.6	5.6
Dar es Salaam	Rainfall	196308	55	–	–	–	–	–	–	–	–	–	–	–	–	–	1.3	15.6
Dar es Salaam	Rainfall	196310	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.3	4.4
Dar es Salaam	Rainfall	196311	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.9	−37.2
Dar es Salaam	Rainfall	196312	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.3	−6.9
Dar es Salaam	Rainfall	196401	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.3	−8.1
Daar es Salaam	Rainfall	196402	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.1	−20.5
Dar es Salaam	Rainfall	196403	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.9	−18.6
Dar es Salaam	Rainfall	196405	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.9	−14.9
Dar es Salaam	Rainfall	196409	55	–	–	–	–	–	–	–	–	–	–	–	–	–	1.1	18.7
Dar es Salaam	Rainfall	196410	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.3	−4.4
Dar es Salaam	Rainfall	196412	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.9	−3.8
Dar es Salaam	Rainfall	196501	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.6	8.1
Dar es Salaam	Rainfall	196502	55	–	–	–	–	–	–	–	–	–	–	–	–	–	1.6	16.2
Dar es Salaam	Rainfall	196503	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.6	−16.8
Dar es Salaam	Rainfall	196504	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.3	−20.5
Dar es Salaam	Rainfall	196505	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.5	−11.2
Dar es Salaam	Rainfall	196506	55	–	–	–	–	–	–	–	–	–	–	–	–	–	0.5	9.9
Dar es Salaam	Rainfall	196507	55	–	–	–	–	–	–	–	–	–	–	–	–	–	2.6	18
Dar es Salaam	Rainfall	196508	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1	1.8
Dar es Salaam	Rainfall	196509	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1	−0.6
Dar es Salaam	Rainfall	196510	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.7	8.7
Dar es Salaam	Rainfall	196511	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.7	−27.3
Dar es Salaam	Rainfall	196512	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.9	−5
Dar es Salaam	Rainfall	196601	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.2	−23.6
Dar es Salaam	Rainfall	196602	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.4	−6.9

(*continued*)

Table 2 | continued

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dar es Salaam	Rainfall	196603	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.8	−23.6
Dar es Salaam	Rainfall	196604	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.5	−10
Dar es Salaam	Rainfall	196605	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.8	−6.9
Dar es Salaam	Rainfall	196606	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.7	−0.6
Dar es Salaam	Rainfall	196607	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.3	−2.5
Dar es Salaam	Rainfall	196608	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.5	−1.3
Dar es Salaam	Rainfall	196609	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.9	7.5
Dar es Salaam	Rainfall	196610	55	–	–	–	–	–	–	–	–	–	–	–	–	–	0.4	13.1
Daar es Salaam	Rainfall	196612	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.3	1.8
Dar es Salaam	Rainfall	196701	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.49	−5.9
Dar es Salaam	Rainfall	196702	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.58	−3.9
Dar es Salaam	Rainfall	196703	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.87	−2.6
Dar es Salaam	Rainfall	196704	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.27	−14.5
Dar es Salaam	Rainfall	196705	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.99	−7.8
Dar es Salaam	Rainfall	196706	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.05	1.4
Dar es Salaam	Rainfall	196707	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.75	−6.5
Dar es Salaam	Rainfall	196708	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.99	3.4
Dar es Salaam	Rainfall	196709	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.76	−3.9
Dar es Salaam	Rainfall	196710	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.52	−11.8
Dar es Salaam	Rainfall	196711	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.79	−24.3
Dar es Salaam	Rainfall	196712	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.76	−11.1
Dar es Salaam	Rainfall	196801	55	–	–	–	–	–	–	–	–	–	–	–	–	–	0.96	10
Dar es Salaam	Rainfall	196802	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.85	−20.6
Dar es Salaam	Rainfall	196803	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−6.68	−37.3
Dar es Salaam	Rainfall	196804	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.94	−25.4
Dar es Salaam	Rainfall	196805	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.66	4
Dar es Salaam	Rainfall	196806	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.28	4.6
Dar es Salaam	Rainfall	196808	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.09	−13.4
Dar es Salaam	Rainfall	196809	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.72	−15.2
Dar es Salaam	Rainfall	196810	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.83	−10.4
Dar es Salaam	Rainfall	196811	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.78	−8

(*continued*)

Table 2 | continued

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^+ (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dar es Salaam	Rainfall	196812	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.29	−17
Dar es Salaam	Rainfall	196901	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.52	−22.4
Dar es Salaam	Rainfall	196902	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.99	−17.7
Dar es Salaam	Rainfall	196903	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.34	−10.6
Dar es Salaam	Rainfall	196904	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.67	−17.7
Daar es Salaam	Rainfall	196905	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.61	−16.3
Dar es Salaam	Rainfall	196906	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.01	−0.9
Dar es Salaam	Rainfall	196907	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.16	6.2
Dar es Salaam	Rainfall	196908	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.12	−13.1
Dar es Salaam	Rainfall	196909	55	–	–	–	–	–	–	–	–	–	–	–	–	–	0.32	8.1
Dar es Salaam	Rainfall	196910	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.66	−0.2
Dar es Salaam	Rainfall	196911	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.37	−2.1
Dar es Salaam	Rainfall	196912	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−8.95	−62.6
Dar es Salaam	Rainfall	197001	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.72	−10
Dar es Salaam	Rainfall	197002	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.59	1.7
Dar es Salaam	Rainfall	197003	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.39	−9.3
Dar es Salaam	Rainfall	197004	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.48	−22.2
Dar es Salaam	Rainfall	197005	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.78	−13
Dar es Salaam	Rainfall	197006	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.63	3.5
Dar es Salaam	Rainfall	197009	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.5	−0.7
Dar es Salaam	Rainfall	197010	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.66	−1.7
Dar es Salaam	Rainfall	197011	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.61	−10.6
Dar es Salaam	Rainfall	197012	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.48	−20.7
Dar es Salaam	Rainfall	197205	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.73	−6.7
Dar es Salaam	Rainfall	197208	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.98	3.8
Dar es Salaam	Rainfall	197210	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.9	−1.9
Dar es Salaam	Rainfall	197211	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.08	−5.9
Dar es Salaam	Rainfall	197212	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.96	−16.8
Dar es Salaam	Rainfall	197301	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.77	−11.2
Dar es Salaam	Rainfall	197302	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.8	4.8
Dar es Salaam	Rainfall	197303	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.04	−9.9

(*continued*)

Table 2 | continued

Source name	Source type	Sample ID	Altitude (m)	Well depth (m)	pH	Temperature (°C)	EC (mS/m)	Alkalinity (meq/L)	Na^{+} (mg/L)	K^{+} (mg/L)	Mg^{2+} (mg/L)	Ca^{2+} (mg/L)	F^{-} (mg/L)	Cl^{-} (mg/L)	NO_3^{-} (mg/L)	SO_4^{2-} (mg/L)	$\delta^{18}O$ (‰)	δD (‰)
Dar es Salaam	Rainfall	197304	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.98	−19.1
Dar es Salaam	Rainfall	197309	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.26	3.2
Daar es Salaam	Rainfall	197310	55	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.26	4.4
Dodoma	Rainfall	199301	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−5.88	−33.8
Dodoma	Rainfall	199302	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	0.06	11.7
Dodoma	Rainfall	199303	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.61	−11
Dodoma	Rainfall	201401	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.95	3.2
Dodoma	Rainfall	201402	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.8	0.6
Dodoma	Rainfall	201411	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.26	−20.4
Dodoma	Rainfall	201412	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.66	3.2
Dodoma	Rainfall	201501	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	3.36	36.4
Dodoma	Rainfall	201502	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.28	−2.3
Dodoma	Rainfall	201503	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−0.68	1.5
Dodoma	Rainfall	201504	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.15	−1.5
Dodoma	Rainfall	201505	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−3.48	−17.4
Dodoma	Rainfall	201511	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−2.58	−8.1
Dodoma	Rainfall	201512	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−4.49	−21.5
Dodoma	Rainfall	201601	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−5.12	−25.3
Dodoma	Rainfall	201602	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−1.25	10.5
Dodoma	Rainfall	201603	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−5.43	−31.3
Dodoma	Rainfall	201604	1157	–	–	–	–	–	–	–	–	–	–	–	–	–	−10.15	−69.6

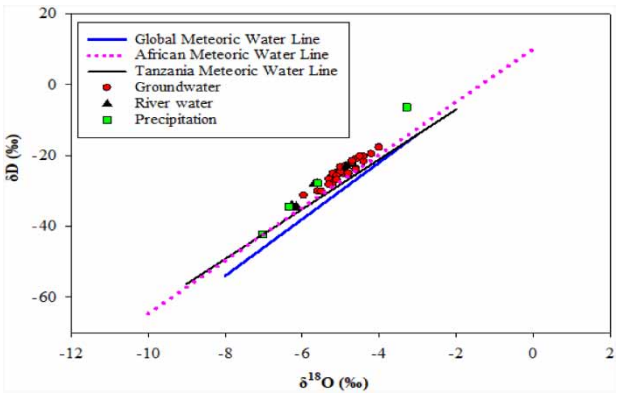

Figure 3 | The plot of δD and δ^{18}O values of groundwater, precipitation and river water in reference to global meteoric water line (GMWL), LMWL and African local meteoric water line.

to the slope of the African meteoric water line (=7.48) and the LMWL for Tanzania (=7.037). However, it is slightly less than the slope of the GMWL (=8), an indication of the minimal influence of evaporative enrichment of precipitation during or prior to infiltration. Moreover, most of the groundwater data plotted close to the LMWL for Tanzania and the African meteoric water line, and according to Farid *et al.* (2014), this indicates that the groundwater in the study area is of meteoric origin (rain and/or snow).

The δ^{18}O of groundwater showed a negative correlation ($r = -0.73$, $P < 0.0004$) with well depth (Table 1b). This suggests the mixing of shallow groundwater with evaporated surface water, for example, from pools stagnated on the land surface in low-lying areas or other fractionated surface water sources. Similar observations were reported in related studies (Gonfiantini *et al.* 1998; Kim *et al.* 2003; Krishnaraj *et al.* 2011). Moreover, the isotopic signature of water samples from wells with depths ≤100 m, particularly samples WG4, WG17, and SP3 with NO_3^- >50 mg/L and high Cl^- concentrations of 34.5, 64.4, and 36.2 mg/L, respectively, suggests recharge by stagnated water pools in low-lying areas or pollution from human activities. These observations agree with previous reports by Olaka *et al.* (2016) and Mduma *et al.* (2016) in related studies.

Groundwater recharge area

The recharge locations of most isotopically depleted groundwater samples (−6.0‰ to −5.0‰) collected from Ngaramtoni area were found at elevations ranging between 2,600 and 3,500 m.a.s.l. The elevation of the recharge areas for most enriched groundwater samples (−4.86‰ to −4.0‰) from the same area, ranged between 1,800 and 2,500 m.a.s.l. in Mount Meru watershed. This indicates that the recharge zone of groundwater used in Ngaramtoni area is virtually similar to that for well water supplied to Arusha city.

The isotopic signature further showed that about 50% of 14 groundwater samples collected at Ngaramtoni area come from a recharge altitude located between 1,800 and 2,500 m.a.s.l. on the south-western slope of Mount Meru. Additionally, more than 60% of 18 groundwater samples from wells used in Arusha city are recharged at an elevation ranging from 2,500 and 3,500 m.a.s.l. on the southern slopes of Mount Meru. This suggests that the well and spring waters used in Arusha urban originate from a recharge area located at altitudes between 1,800 and 3,500 m.a.s.l. in Mount Meru watershed. Well depth showed a significant positive relationship ($r = 0.73$, $P < 0.05$) with recharge elevation (Table 1b). This can be an indication that shallow groundwater comes from low altitude recharge areas while deep groundwater originates from high altitude recharge areas.

Water sample WG4 collected from an artesian well (100 m deep) was observed to have high altitude isotopic signature (-5.0‰), and elevated NO_3^- concentration (83.5 mg/L), which indicates pollution due to human activities in the recharge area. Carrillo-Rivera & Varsányi (2007) also reported similarly in a related study. The mean fluoride value of 6.1 ± 0.49 mg/L was observed in groundwater coming from high-altitude recharge areas between 2,537 and 3,500 m.a.s.l. On the other hand, groundwaters originating from low altitude recharge areas between 1,800 and 2,530 m.a.s.l. was observed to have relatively low average fluoride content (4.3 ± 0.5 mg/L), suggesting that the fluorite precipitation CaF2 at the lower altitude reduces the fluoride contents in the groundwater system (Edmunds & Smedley 2013).

The water sample from spring (SP3) located at a high altitude (1,582 m.a.s.l.) was observed to have an elevated fluoride concentration (5.5 mg/L) and enriched isotopic values (−4.6‰). According to Plummer *et al.* (2001), this indicates the mixing of waters of variable ages and quality along the flow pathways. Sample WG17 was collected in a shallow well (48 m deep) and appeared to originate from a mixture

of groundwater and contaminated surface water with negligible fluoride contents. This was depicted by the elevated NO_3^- contents (83.9 mg/L), enrichment in isotopic values of oxygen (−4.2‰), and low fluoride concentration (2.2 mg/L). Therefore, infiltration characteristics and the duration that water stays on the land surface at different locations in the topographically lower altitudes show some relationship between $\delta^{18}O$ and NO_3^- concentration of groundwater.

Groundwater flow path

Groundwater samples collected from the entire study area were plotted for fluoride contents versus the recharge elevations (Figure 4(a)), which conceptually indicated two groups of water quality (A and B) based on the relationship between fluoride concentration and the stable isotopic ratio of oxygen of groundwater samples (Figure 4(d)). Similarly, from the data, it can be inferred that there are two different flow pathways from two recharge areas. The fluoride contents in water quality group A ranged from 5.2 to 10.1 mg/L with a mean value of 7.2 ± 0.5 mg/L, while for the group B, the range was from 1.2 to 7.3 mg/L with a mean value of 4.2 ± 0.4 mg/L. Those suggest a difference in locations, geology, or catchment areas of the groundwater flow pathways in the study area. The concentration of fluoride in the two flow pathways was shown to increase with well depth (Figure 4(b)). The fluoride contamination in the flow pathway of water group A showed a poor relationship with Na^+ concentration and a positive significant correlation in group B (Figure 4(c)). This indicates that the fluoride is dissolved through water–rock interaction over a longer period with weathering of granite along the flow pathway or in the recharge areas of water group B (Kim & Kim 2011).

The isotopic ratio of oxygen showed an increasing trend as the fluoride contamination was reduced in all flow pathways (Figure 4(d)). Similar cases have been reported in Tanzania and around the globe (Mduma *et al.* 2016; Olaka *et al.* 2016), suggesting the mixing of high fluoride-contaminated groundwater from topographically high areas and shallow groundwater from local recharge or polluted surface runoff water pools stagnated in low-lying areas with negligible fluoride contents along the flow direction.

Figure 1 provides the details of three locations (1, 2, and 3) of the discharge areas of the two flow pathways. Comparing the fluoride contents in both flow pathways, the high fluoride leaching at location 1 (mean value = 6.3 ± 0.5 mg/L) is an indication of the mixing of water with relatively high fluoride contamination from the two flow pathways as presented in Figure 1. Moreover, Figure 1 shows the low concentration of fluoride at location 3 (mean value = 4.3 ± 0.7 mg/L), suggesting the dilution of fluoride contaminated water from the two flow pathways with water of negligible fluoride contents like rainwater stagnated in low-lying areas.

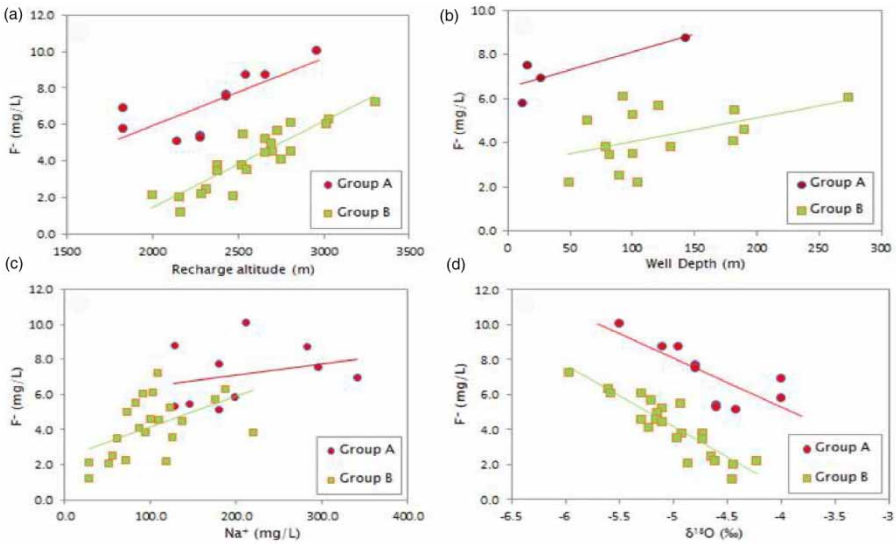

Figure 4 | Variation of F^- concentration with recharge altitude (a), well depth (b), with Na^+ (c), and isotopic composition of groundwater (d).

The concentration of NO_3^- at location 1 varied between 4.6 and 77.0 mg/L with a mean value of 22.4 ± 5.8 mg/L. The groundwater at location 2 was observed to have relatively low pollution of NO_3^-, as the measured value ranged between 0.7 and 37.8 mg/L with a mean value of 11.9 ± 3.2 mg/L. A relatively wide range of NO_3^- contamination ranging between 1.9 and 83.9 mg/L with a mean value of 32.6 ± 10.6 mg/L was observed in the groundwater collected at location 3, indicating contamination from anthropogenic sources. This is similar to the reports by Makoba & Muzuka (2019), Mduma *et al.* (2016), and Elisante & Muzuka (2016) on groundwater sources in the slopes of Mount Meru.

CONCLUSIONS AND RECOMMENDATIONS

The altitude effect of the isotopic ratio of oxygen revealed that groundwater used in Arusha urban, northern Tanzania, comes from recharge areas located at altitudes between 1,800 and 3,500 m.a.s.l. The groundwater in the study area is of meteoric origin (rain and/or snow). The isotopic signature and the spatial distribution of fluoride contamination in the groundwaters generally indicate two flow pathways which start from the recharge area in the south and south-western slopes of Mount Meru toward the southern part of Arusha urban.

Relatively lower fluoride levels and elevated NO_3^- in groundwater from the low-lying parts of the study area could be due to dilution by local recharge with negligible fluoride contents. The contamination of fluoride and NO_3^- at some sampling locations confirm that the low-altitude groundwater comes from high-altitude and local recharge sources and the high-altitude groundwater is recharged from topographically high areas of Mount Meru. Elevation of NO_3^- and Cl^- contents in water samples from sources in the lower part of the study area is the evidence of anthropogenic contamination.

The fluoride concentration in the studied groundwaters is natural contamination and exceeds the WHO and Tanzania's guidelines of 1.5 mg/L by 70%. The fluoride concentrations in the river waters similarly exceeded the standard by 75%. The fluoride contamination seems to increase toward the recharge areas and there is evidence of mixing of high fluoride groundwater from the two flow pathways. Dilution effects lowered the concentration of fluoride in groundwater in the lower part of the study area. This study, therefore, recommends (1) protection of the local groundwater recharge areas from anthropogenic contamination as they produce water with relatively low fluoride contents and (2) application of appropriate treatment method to moderate the fluoride concentration in the water supplied to Arusha urban.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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